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Radiological Aspects of Deep-Burn Fusion-Fission Hybrid Waste in a Repository

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ABSTRACT

The quantity, radioactivity, and isotopic characteristics of the spent fission fuel from a hybrid fusion-fission system capable of extremely high burnups are described. The waste generally has higher activity per unit mass of heavy metal, but much lower activity per unit energy generated. The very long-term radioactivity is dominated by fission products. Simple scaling calculations suggest that the dose from a repository containing such waste would be dominated by ^{129}I, ^{135}Cs, and ^{242}Pu. Use of such a system for generating energy would greatly reduce the need for repository capacity

INTRODUCTION

Lawrence Livermore National Laboratory is developing a hybrid fusion-fission nuclear energy system, called LIFE (Laser Inertial-confinement Fusion-fission Engine) to generate electrical power and burn nuclear waste [1, 2]. The system uses the neutrons produced by laser-driven inertial confinement fusion to produce additional fusion fuel and to drive nuclear reactions in a subcritical fission blanket. The fusion neutron source obviates the need for a self-sustaining chain reaction in the fission blanket. Both fissile fuels (*e.g.*, ^{235}U, ^{239}Pu), and fertile fuels (*e.g.*, depleted uranium, natural uranium, ^{232}Th, or spent LWR fuel) could be used as fission fuel, thus eliminating the need for isotopic enrichment. The fusion neutrons allow extremely high levels of burnup to be reached, extracting a large fraction of the available energy in the fission fuel without the need for reprocessing. In this paper, we discuss the radionuclide inventory of a depleted uranium (DU) fuel burned to greater than 95 % FIMA (Fissions per Initial heavy Metal Atom), and the implications of the resulting waste relative to dose standards for releases from a geological repository for high-level waste.

The LIFE system producing the waste discussed here uses low-yield (37.5 MJ), deuterium-tritium fusion targets ignited at a rate of 13.3 Hz to produce a 500 MW fusion source. Of this energy, ~400 MW is carried of by 14 MeV neutrons (1.8×10^{20} n/s), and the remaining energy is carried off in ions and x-rays. The fusion neutrons are multiplied and moderated by a sequence of concentric shells of materials before encountering the fission fuel. The fission blanket contains, in this case, 40 metric tons (MT) of DU fuel. For this analysis, the fuel was assumed to be TRISO-like UOC fuel particles embedded in 2-cm-diameter graphite pebbles. (Present TRISO fuel designs may not reach of the high burnups of the fertile fuel considered here, and other fuel options, including molten salt fuel, are being investigated. Here, we assume the existence of a fuel that can reach >95% FIMA.) The fission fuel pebbles are cooled by a molten LiF-BeF_2 (flibe) coolant, which also produces tritium for the fusion portion of the engine. The flibe is pumped through the fission blanket and sent to heat exchangers to drive a Brayton-cycle power plant. The engine and plant design used here would receive one load of fission fuel and produce ~2 MWt of power over its 50- to 70-year lifetime.

CALCULATIONS

Neutron and photon transport calculations were performed using the Monte Carlo transport code MCNP5 [3]. Burnup calculations were performed using Monteburns 2.0 [3], which, in turn, uses ORIGEN2 [4] for depletion/decay calculations. Improvements to Monteburns, as well as additional custom code developments, were required to perform the burnup calculations for LIFE. The nuclear data used were from ENDF/B-VII [6]. Additional details of the burn calculations can be found in [2].

For comparison to spent fuel from light water reactors (LWRs), we use the projected initial inventory of PWR and BWR fuels (average age of 23 years since discharge) used for the Yucca Mountain Project Final Environmental Impact Statement [7]. The evolution of this initial inventory to 1 million years was calculated using ORIGEN2.

DISCUSSION

Waste quantities

The LIFE plant design used here would generate approximately 44 GWe-yr of energy when the fuel reaches 99% FIMA [2], and because the engine was fueled with 40 MT of DU, the energy generated per MT is about 1.1 GWe-yr/MT. In contrast, using average burnups of 41.2GWt-day/MT and 33.6GWt-day/MT for the PWR and BWR fuel slated for disposal at Yucca Mtn. [7], and assuming a thermal-electric conversion efficiency of ~33%, the total electricity generated by the 68,000 MT of spent fuel slated for disposal at Yucca Mountain is ~2500 GWe-yr, or ~3.9×10^{-2} GWe-yr/MT, which is about 28 times less energy per MT than the LIFE waste. Clearly, relative to the current once-through fuel cycle, the use of LIFE engines to generate electricity would significantly reduce the need for repository capacity.

Radiological characteristics and inventory

The specific radioactivity (curies/MT) of spent LIFE fuel as a function of time after discharge is shown in Figure 1a. For the first ~300 years, the specific activity for 95%, 99%, and 99.9% FIMA is significantly higher than that of average LWR fuel. The specific activity of LIFE waste with a burnup of 95% FIMA remains above that of average LWR fuel for all times. LIFE waste with a burnup of 99% FIMA has a specific activity similar to that of average spent LWR fuel up between ~300 years to ~100,000 years post discharge, while the 99.9% FIMA waste has a specific activity less than that of average LWR fuel from ~300 years to 100,000 years post discharge. At very long times (>300,000 years), the specific activities of the spent LIFE fuels for all three burnups are somewhat higher than that of average spent LWR fuel.

Figure 1b shows the same data normalized to the total electrical energy generated. It is clear from Figure 1b that the radioactivity per-unit-energy-generated of spent LIFE fuel is less than that of similarly normalized spent LWR fuel, suggesting that the benefit-to-hazard ratio of LIFE waste is significantly better than that of spent LWR fuel. Nevertheless, the spent fission fuel from a LIFE engine is a hazardous material that would require isolation from the biosphere for at least hundreds of thousands of years.

For decay times of less than ~300 years, the activity of spent LIFE fuel is dominated by short-lived fission products. Specifically, the activity of the waste (regardless of burnup) is dominated by the decay of ^{137}Cs + ^{137m}Ba, and ^{90}Sr + ^{90}Y for the first few hundred years after

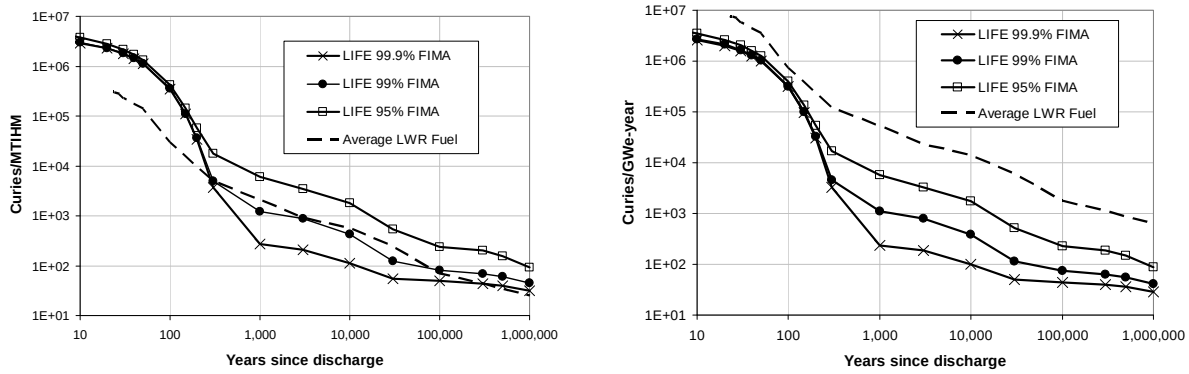

Figure 1. (a) Comparison of the specific activity of spent LIFE fuel as a function of time since discharge with that of average LWR fuel. **(b)** Activity of spent LIFE fuel normalized to the total energy generated by that fuel. For comparison, the activity of average LWR fuel is also plotted.

discharge (Figure 2a). These are the same nuclides responsible for most of the activity of spent LWR fuel during the same time period (though the specific activity of LWR fuel is lower).

Between ~300 and a few tens of thousands of years (Figure 2b), decay of the actinides and their daughter products (^{246}Cm, ^{245}Cm+daughters, ^{240}Pu) are the dominant sources of radioactivity in spent LIFE fuel. At times greater than ~20,000 years, fission products (^{135}Cs, ^{93}Zr + ^{93m}Nb and ^{99}Tc) again become the dominant radioactivities. ^{242}Pu is the only actinide that contributes more than 5% of the total activity during the post-100,000-year time period. The radioisotopic makeup of LIFE waste differs substantially from average LWR spent fuel; the long-term activity in spent LWR fuel is dominated by the decay of ^{99}Tc, ^{241}Am, ^{239}Pu, ^{240}Pu, ^{242}Pu, ^{226}Ra+daughters, and ^{229}Th+ daughters.

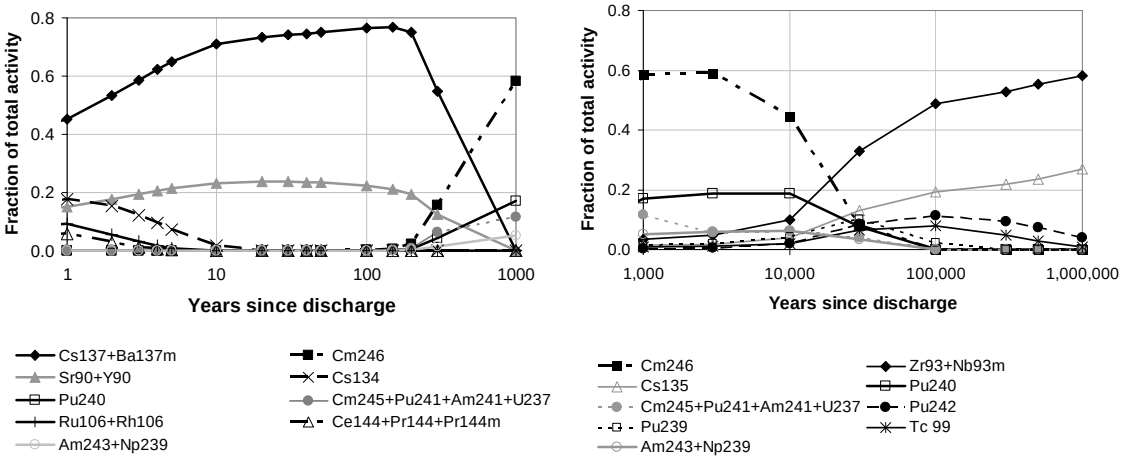

Figure 2. (a) Fractional contribution of individual nuclides and decay chains to the total activity of 99% FIMA LIFE fuel for 1 to 1000 years post discharge. (b) Fractional contribution of individual nuclides and decay chains to the total activity of 99% FIMA LIFE fuel for 1000 to 1,000,000 years post discharge. (Figure includes all nuclides or chains that contribute more than 5% to the total activity at any time during these periods.)

The actinide concentration in LIFE fuel as a function of burnup is given in Table 1. From 20% to ~60% FIMA, LIFE fuel contains roughly 10 times the concentration of transuranic elements (TRU) than does average LWR fuel at the time of discharge. This is similar to other high-burnup advanced fuel cycle options. With increasing burnup, these elements begin to "burn out", and by 95% FIMA, their concentration is approximately twice that in average spent LWR fuel. By 99% FIMA, they are at a concentration half that of LWR fuel, and at 99.9% FIMA, they are about a tenth the LWR concentration. (Note that unlike spent LWR fuel, most of the initial uranium has also been transmuted in LIFE waste, so the total mass of actinides is much less.) Comparing the production of TRU as a function of the energy produced by a LIFE engine vs. a conventional LWR with no reprocessing, the LIFE engine produces less TRU per unit energy generated for all burnups greater than ~50%. A LIFE engine operating to a burnup of 99% FIMA would produce ~60 times less TRU waste than an average LWR per unit energy generated.

Table 1. Uranium and transuranic (TRU) element content of LIFE fuel as a function of burnup. Data are also provided for average spent LWR fuel.

	Kg per MT initial U at discharge at indicated burnup						Kg per GWe-year at discharge at indicated burnup					
% FIMA	60%	80%	95%	99%	99.9%	avg. LWR	60%	80%	95%	99%	99.9%	avg. LWR
U	243	106	24	4.5	0.21	955	366	121	23	4.1	0.18	24476
Np	0.91	0.47	0.16	0.039	0.007	0.68	1.4	0.53	0.15	0.035	0.006	17
Pu	135	69	12	0.7	0.02	10	203	79	11	0.65	0.018	254
Am	10	7.3	1.8	0.17	0.004	1.2	15.3	8.3	1.7	0.16	0.004	30
Cm	13	16	11	4.4	1.0	0.03	19	18	11	4.0	0.93	0.69
Bk	0.008	0.021	0.026	0.016	0.001		0.012	0.023	0.025	0.014	0.001	
Cf	0.026	0.054	0.072	0.100	0.010		0.039	0.061	0.068	0.090	0.009	
Total TRU	158	93	25	5.4	1.1	12	239	106	24	4.9	0.97	302

Implications for dose from a repository

The proposed U.S. Nuclear Regulatory Commission Rule (10 CFR 63.311) [8, 9] for licensing the Yucca Mountain repository requires that there is a reasonable expectation that the maximally exposed individual would receive a dose of <15 mrem/yr from releases from the undisturbed repository system for the first 10,000 years after disposal, and<350 mrem/yr between 10,000 and 1,000,000 years. Results from the Yucca Mountain Total System Performance Assessment (TSPA) indicate that the proposed repository design will meet these requirements [10]. The TSPA results for show that ^{14}C, ^{99}Tc, ^{129}I, and ^{239}Pu dominate the dose for the first 10,000 years after repository closure. ^{239}Pu, ^{129}I, and ^{226}Ra dominate the dose for the next 90,000 years, and ^{226}Ra, ^{237}Np, and ^{242}Pu, dominate the dose from 100,000 to one-million years. Other radionuclides are significant under specific failure scenarios.

Figure 3 shows the ratios of the specific activities of these radionuclides in LIFE waste (99% FIMA) to their values for average LWR fuel. The figure includes several other radionuclides (^{93}Zr, ^{244}Pu, ^{246}Cm, ^{248}Cm) that may be important for LIFE waste, but were not considered in the Yucca Mountain TSPA because they were deemed unimportant. With few exceptions, the specific activity of the actinides and daughter products are much lower in LIFE waste than in average LWR fuel; LIFE waste has higher activities of ^{242}Pu, ^{244}Pu, ^{246}Cm, and ^{248}Cm. Among the fission products of concern, ^{99}Tc and ^{126}Sn have lower specific activities in

LIFE waste, while ^{129}I, ^{135}Cs, and ^{93}Zr have higher specific activities. When normalized to the energy produced, however, only ^{246}Cm has higher abundance in LIFE waste.

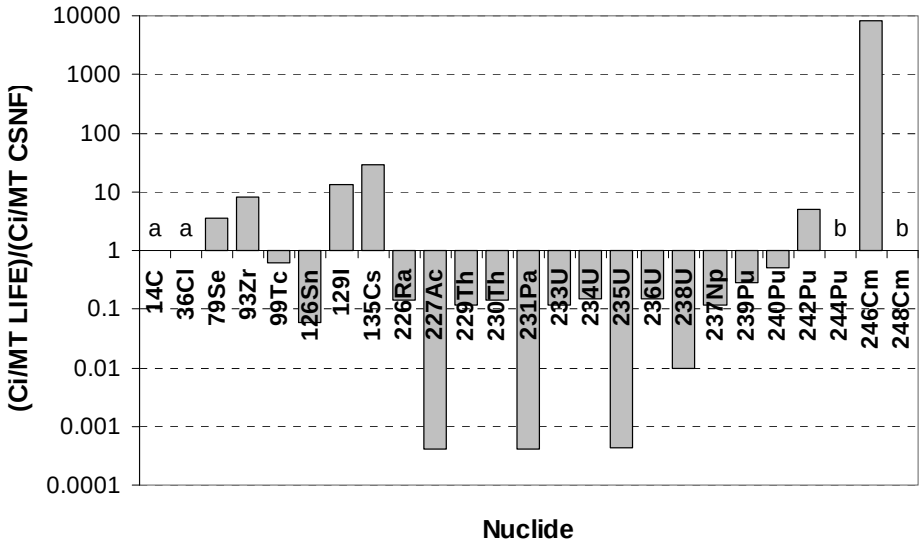

Figure 4. Ratios of the specific activities of radionuclides important for long-term repository performance in spent LIFE fuel (99% FIMA) to their values for average spent LWR fuel. Notes: (a) data on ^{14}C and ^{36}Cl are not available for LIFE waste due to limitations in the nuclear cross section data used; (b) ^{242}Pu and ^{248}Cm are not present in significant quantities in spent LWR fuel.

By making the simplifying assumption that the dose from a given radionuclide is proportional to its inventory, one can make a first-order assessment of the impact of replacing the spent LWR fuel in a repository with LIFE waste. The following additional assumptions and approximations were made or are implicit in this method:

- YMP TSPA doses results are dominated by the CSNF inventory.
- LIFE waste packages and fuel degrade and release radionuclides at the same average rate as the waste considered in the YMP TSPA-LA
- ^{246}Cm and ^{248}Cm are accounted for by their daughters, ^{242}Pu and ^{244}Pu activity [10,11]
- ^{244}Pu and ^{242}Pu have equal dose-conversion factors, and ^{244}Pu doses were scaled to the ratio of ^{244}Pu and ^{242}Pu
- ^{93}Zr is released and transported at the same rate as ^{229}Th.

This assessment is only valid for the post ~25,000-year performance period. A similar assessment for times < 25,000 years cannot be done because we are lacking data on the ^{14}C content of LIFE waste due to limitations in the cross-section data used for the burn calculations.

The results indicate that the doses from a "LIFE repository" containing the same mass of initial heavy metal would be 4-5 times higher than the YMP TSPA-LA results for this time period, though the waste in the repository would have produced ~28 times the energy. The doses do not come close to approaching the proposed NRC limit of 350 mrem/yr. Most of the dose comes from ^{129}I, ^{135}Cs, and ^{242}Pu. Although the specific activity of ^{244}Pu is much higher in LIFE waste than in spent LWR fuel, the absolute amount of ^{244}Pu is small, and it does not appear that it will be a major dose contributor. Similarly, although ^{93}Zr+^{93m}Nb are the dominant activities at long times, they would not contribute substantially to dose due to their low dose-conversion

factors. Note that this calculation provides insight into which radionuclides in LIFE waste would be likely to contribute significantly to the dose from a repository similar to the proposed repository at Yucca Mtn.; other geologic settings or repository designs could have significantly different results.

CONCLUSIONS

Use of a fusion-fission hybrid system would allow high burnups to be achieved in a subcritical fission blanket. Per unit energy generated, the quantity of spent fission fuel from such a system could be a factor of 25-30 less than for the current once-through fuel cycle. The dose from a repository containing such waste would likely be dominated by ^{129}I, ^{135}Cs, and ^{242}Pu.

ACKNOWLEDGMENTS

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